# Supporting Information

# High Temperature Thermoelectric Properties of the Solid-Solution Zintl Phase $Eu_{11}Cd_{6-x}Zn_xSb_{12}$

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To investigate the quality of each sample after hot pressing, synchrotron powder XRD (SPXRD) was utilized. Representative patterns of the $x_p$ = 1, 2, 5, and 6 samples of $Eu_{11}Cd_{6-x}Zn_xSb_{12}$ solid solutions are shown in Figure S1. The best profile matching was achieved by using the crystal structures solved for crystals of each reaction. The corresponding calculated patterns are shown as orange dashed lines overlaid on each observed pattern, shown in black. This profile matching results in the best figures of merit when the main phase with 11-6-12 structure and the minor phase related to the 9-4-9 structure were used in refinement. Samples with $x_p$ = 3 and 4 showed high volume fractions of 9-4-9 structure resulting to not be considered in this study. Lebail profile matching on the powder patterns of the other samples with $x_p$ = 1, 2, 5, and 6 showed a minor contribution from 9-4-9 structure.

Zn Lα X-ray maps given by EMPA on single crystals and pressed pellets are provided in Figure S2. The average compositions with standard deviations in parentheses are also listed below the corresponding images. There is good agreement between the compositions obtained from single crystal X-ray diffraction and from EMPA. Zn distribution on all samples show homogenous samples except in $x_p$ = 2 pellet that two regions of high Zn and low Zn contents are making larger standard deviations.

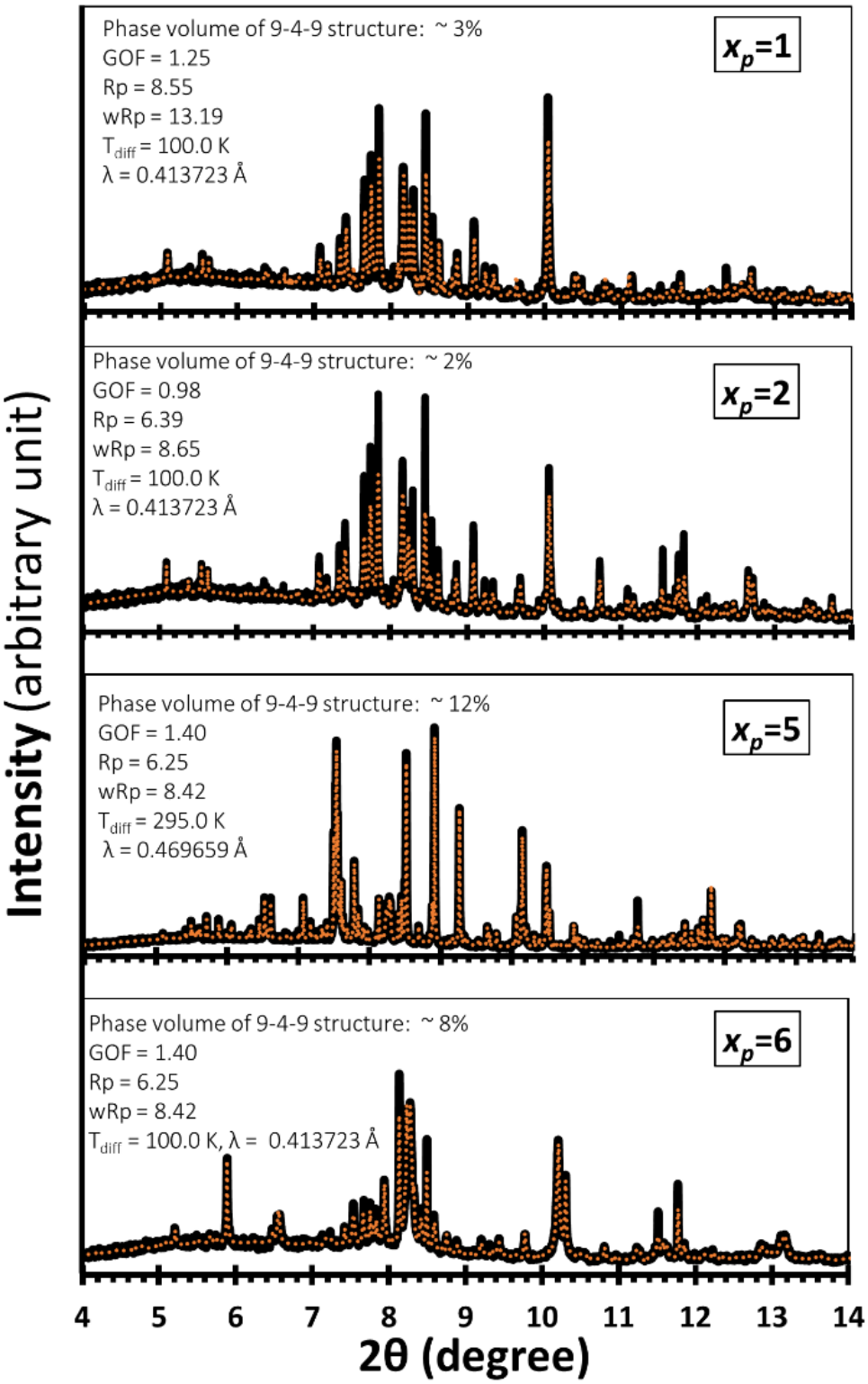


**Figure S1.** High resolution synchrotron powder X-ray diffraction patterns for $Eu_{11}Cd_{6-x}Zn_xSb_{12}$ solid solutions ($x_p$ = 1, 2, 5, and 6) and their overlaid whole profile refinement (LeBail method) patterns (in orange dashed lines). Their corresponding statistical figures of merit of GOF, $R_p$, and $wR_p$ along with their experimental conditions are listed for each pattern.

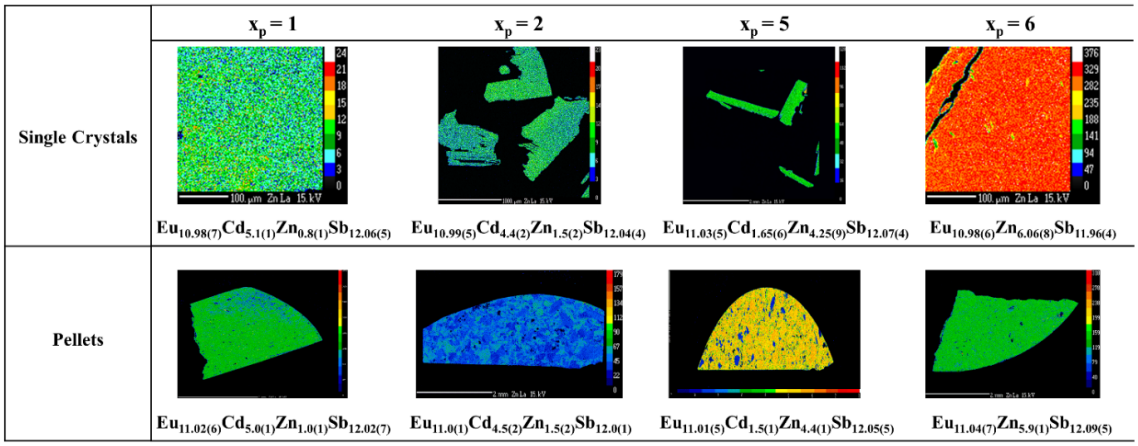


**Figure S2.** Zn Lα X-ray maps of single crystals and pressed slices for $Eu_{11}Cd_{6-x}Zn_xSb_{12}$ solid solutions ($x_p$ = 1, 2, 5, and 6) and their corresponding average compositions are shown.

**Table S1.** Thermal Diffusivity, Heat Capacity and Density Data for $Eu_{11}Cd_{6-x}Zn_xSb_{12}$ solid solutions ($x_p$ = 1, 2, 5, and 6)

| #Database | | | Cp (J/g*K) | 0.19612276 |
|---|---|---|---|---|
| #Instrument | | #LFA_457 | density | 6.460 |
| #Sample | | x = 4.4 | | |
| ##Results | | | | |
| temp © | temp (K) | fusivity(mm^2 | d_Dev (mm^2/ | kappa |
| 50.7 | 323.7 | 0.797 | 0.002 | 1.009800398 |
| 100.6 | 373.6 | 0.769 | 0.002 | 0.974324349 |
| 150.8 | 423.8 | 0.74 | 0.002 | 0.937581298 |
| 200.9 | 473.9 | 0.639 | 0 | 0.809614121 |
| 250.8 | 523.8 | 0.635 | 0.001 | 0.804546114 |
| 300.9 | 573.9 | 0.617 | 0 | 0.781740082 |
| 350.9 | 623.9 | 0.599 | 0.001 | 0.758934051 |
| 401 | 674 | 0.582 | 0.002 | 0.737395021 |
| 451 | 724 | 0.586 | 0 | 0.742463028 |
| 501.1 | 774.1 | 0.616 | 0.001 | 0.780473081 |
| 375.4 | 648.4 | 0.573 | 0.001 | 0.725992005 |
| 275.4 | 548.4 | 0.574 | 0.001 | 0.727259007 |
| 175.5 | 448.5 | 0.607 | 0.001 | 0.769070065 |
| 75.7 | 348.7 | 0.661 | 0.002 | 0.83748816 |

| | | | Cp (J/g*K) | 0.20020759 |
|---|---|---|---|---|
| #Instrument | | #LFA_457 | density | 6.552 |
| #Sample | | x = 6 | | |
| ##Results | | | | |
| temp (C) | temp (K) | fusivity(mm^2 | d_Dev (mm^2/ | kappa |
| 50.6 | 323.6 | 0.756 | 0.001 | 0.991763958 |
| 100.5 | 373.5 | 0.701 | 0.001 | 0.919611818 |
| 150.6 | 423.6 | 0.657 | 0.001 | 0.861890106 |
| 200.8 | 473.8 | 0.611 | 0.002 | 0.80154468 |
| 250.9 | 523.9 | 0.574 | 0.001 | 0.753005968 |
| 300.9 | 573.9 | 0.563 | 0.001 | 0.73857554 |
| 350.9 | 623.9 | 0.563 | 0.001 | 0.73857554 |
| 400.9 | 673.9 | 0.569 | 0 | 0.746446682 |
| 450.9 | 723.9 | 0.591 | 0.001 | 0.775307538 |
| 500.9 | 773.9 | 0.624 | 0 | 0.818598822 |
| 300.2 | 573.2 | 0.562 | 0.001 | 0.737263683 |
| 100.6 | 373.6 | 0.677 | 0.001 | 0.888127248 |
| 31 | 304 | 0.758 | 0.002 | 0.994387672 |

| #Database | | | Cp (J/g*K) | 0.1891287 |
|---|---|---|---|---|
| #Instrument | | #LFA_457 | density | 6.380 |
| #Sample | | x = 1.5 | | |
| ##Results | | | | |
| temp (C) | temp (K) | fusivity(mm^2 | d_Dev (mm^2/ | kappa |
| 46.3 | 319.3 | 0.738 | 0.005 | 0.890540367 |
| 99.5 | 372.5 | 0.698 | 0.002 | 0.842272597 |
| 148.7 | 421.7 | 0.661 | 0 | 0.797624909 |
| 200.9 | 473.9 | 0.626 | 0.001 | 0.75539061 |
| 252.9 | 525.9 | 0.585 | 0.001 | 0.705916145 |
| 302.6 | 575.6 | 0.571 | 0.001 | 0.689022425 |
| 352.2 | 625.2 | 0.567 | 0.001 | 0.684195648 |
| 401.9 | 674.9 | 0.558 | 0.001 | 0.6733354 |
| 451.6 | 724.6 | 0.55 | 0.002 | 0.663681846 |
| 501.5 | 774.5 | 0.541 | 0.001 | 0.652821597 |
| 298 | 571 | 0.55 | 0.001 | 0.663681846 |
| 99.1 | 372.1 | 0.66 | 0.004 | 0.796418215 |
| 38.9 | 311.9 | 0.709 | 0.005 | 0.855546234 |

| | | | Cp (J/g*K) | 0.18797294 |
|---|---|---|---|---|
| #Instrument | | #LFA_457 | density | 6.660 |
| #Material | | x = 1 | | |
| ##Results | | | | |
| temp (C) | temp (K) | fusivity(mm^2 | d_Dev (mm^2/ | kappa |
| 50.3 | 323.3 | 0.867 | 0.003 | 1.08539712 |
| 100.5 | 373.5 | 0.817 | 0.002 | 1.02280213 |
| 150.6 | 423.6 | 0.778 | 0.001 | 0.973978038 |
| 200.8 | 473.8 | 0.66 | 0.003 | 0.826253863 |
| 250.8 | 523.8 | 0.662 | 0.001 | 0.828757662 |
| 301 | 574 | 0.663 | 0.001 | 0.830009562 |
| 351 | 624 | 0.661 | 0.001 | 0.827505762 |
| 401 | 674 | 0.662 | 0.001 | 0.828757662 |
| 450.9 | 723.9 | 0.666 | 0 | 0.833765261 |
| 500.9 | 773.9 | 0.658 | 0.002 | 0.823750063 |
| 374.6 | 647.6 | 0.64 | 0.002 | 0.801215867 |
| 274.7 | 547.7 | 0.628 | 0.001 | 0.786193069 |
| 174.9 | 447.9 | 0.671 | 0.001 | 0.84002476 |
| 75.3 | 348.3 | 0.747 | 0.002 | 0.935169145 |